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INFORMAL REPORT

LYSIMETER DATA FROM EPICOR-II WASTE FORMS--FISCAL YEAR 1986

Robert D. Rogers
John W. McConnell, Jr.
Melvin W. Findlay
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EG&G Idaho
Work performed under DOE Contract No. DE-AC07-76ID01570

Prepared for the
U.S. NUCLEAR REGULATORY COMMISSION

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ABSTRACT

A field study was designed to monitor the release (if any) of beta- and gamma-producing radionuclides from solidified EPICOR-II ion exchange resins. Both Portland Type I-II cement and Dow vinyl ester-styrene waste forms are being tested in lysimeter arrays located at Argonne National Laboratory in Illinois (ANL-E) and Oak Ridge National Laboratory (ORNL). The study is designed so that continuous data on nuclide release and movement, as well as environmental conditions, will be obtained over a 20-yr period. Results of the first year of data acquisition are presented and discussed. Unusual occurrences over that period are also described.

SUMMARY

One of the tasks developed to address the disposal of resin wastes contained in EPICOR-II prefilters is field testing. The purpose of the task, using lysimeter arrays at Oak Ridge National Laboratory (ORNL) and Argonne National Laboratory in Illinois (ANL-E), is to expose samples of solidified resin waste to the actual physical, chemical, and microbiological conditions of a burial environment. The study is designed so that continuous data on nuclide release and movement, as well as environmental conditions, will be obtained over a 20-yr period.

Lysimeters used in this study were designed to be self-contained units. Each is a right-circular cylinder divided into an upper compartment, which contains fill material, waste forms, and instrumentation, and a lower compartment, which collects leachate. Four lysimeters at each site are filled with soil, while a fifth (used as a control) is filled with inert silica oxide sand. Instrumentation within each lysimeter includes porous cup soil-water samplers and soil moisture/temperature probes. The probes are connected to an on-site data acquisition and storage system (DAS) which also collects data from a field meteorological station located at each site.

Each month, data stored on a cassette tape are retrieved from the DAS and translated into an IBM PC compatible disk file. At least quarterly, water is drawn from the porous cup soil-water samplers and the lysimeter leachate collection compartment. Those water samples are analyzed for beta- and gamma-producing nuclides.

Results of the first year of data acquisition are presented in this report. These results show that radionuclides are moving from the waste forms into the soil column much sooner than expected. Also, soil and precipitation differences have resulted in more ^{90}Sr movement at ORNL than at ANL-E.

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LYSIMETER DATA FROM EPICOR-II WASTE FORMS--FISCAL YEAR 1986

INTRODUCTION

Solidified waste forms containing EPICOR-II ion exchange resin waste are currently being field-tested using lysimeters. The intent of the testing is to expose waste form samples to the physical, chemical, and microbiological environment of typical disposal sites in the eastern United States. It is intended that the lysimeters be used as tools to monitor release of nuclides from the buried waste forms and provide data which will allow for the accurate determination of movement as a function of time and environmental conditions.

Lysimeter sites have been established at Oak Ridge National Laboratory (ORNL) and Argonne National Laboratory-East (ANL-E). Instrumentation within each of the five lysimeters at each site includes porous cup soil-water samplers and soil moisture/temperature probes. The probes are connected to an on-site data acquisition and storage system (DAS) which also collects data from a field meteorological station located at each site. A detailed description of the lysimeters and their installation is contained in an earlier report.[1]

This report contains data from the first year of operation of the lysimeters. These data include information retrieved from the DAS as well as beta and gamma analyses of water which has passed through the lysimeters.

MATERIALS AND METHODS

Description of Waste Forms

Waste forms used in the field test are composed of solidified EPICOR-II prefilter resin wastes. Two waste formulations are used in the solidification project. Type A is a mixture of synthetic organic ion exchange resins from PF-7 (phenolic cation, strong acid cation, and strong base anion resins), while Type B is a mixture of synthetic organic ion exchange resins from PF-20 (strong acid cation and strong base anion resins) with an inorganic zeolite. Portland Type I-II cement and vinyl ester-styrene (a proprietary solidification agent developed and supplied by the Dow Chemical Company) were used to solidify both types of resin wastes. Individual waste forms were manufactured by allowing a mixture of solidification agent and resin waste to solidify in 4.8-cm-diameter by 10.2-cm-high polyethylene molds. Enough of the mixture was added to each vial to produce waste forms with an average dimension of 4.8 by 7.6 cm (137.5 cm^3), as shown in Figure 1. A complete description of waste form manufacture is given in Reference 2, while bench testing of similar waste forms is described in Reference 3.

Description of Lysimeters

The lysimeters are designed as self-contained units which can be easily disposed at the termination of the field test experiment. A total of 10 lysimeters are used, with five placed at each field site. Each lysimeter is a right-circular cylinder (0.91 m ID by 3.12 m in height) constructed ot 12-gage, 316L stainless steel (Figure 2). Internally, the lysimeter is divided into two sections, the upper being 1532 L in volume and the lower being 396 L. A 3.8-cm, Schedule 40, stainless steel pipe serves as an access to the lower compartment. Soil, instrumentation, and waste forms are contained in the upper compartment, while the lower compartment serves as a leachate collector.

Figure 1. An example of an EPICOR-II waste form.

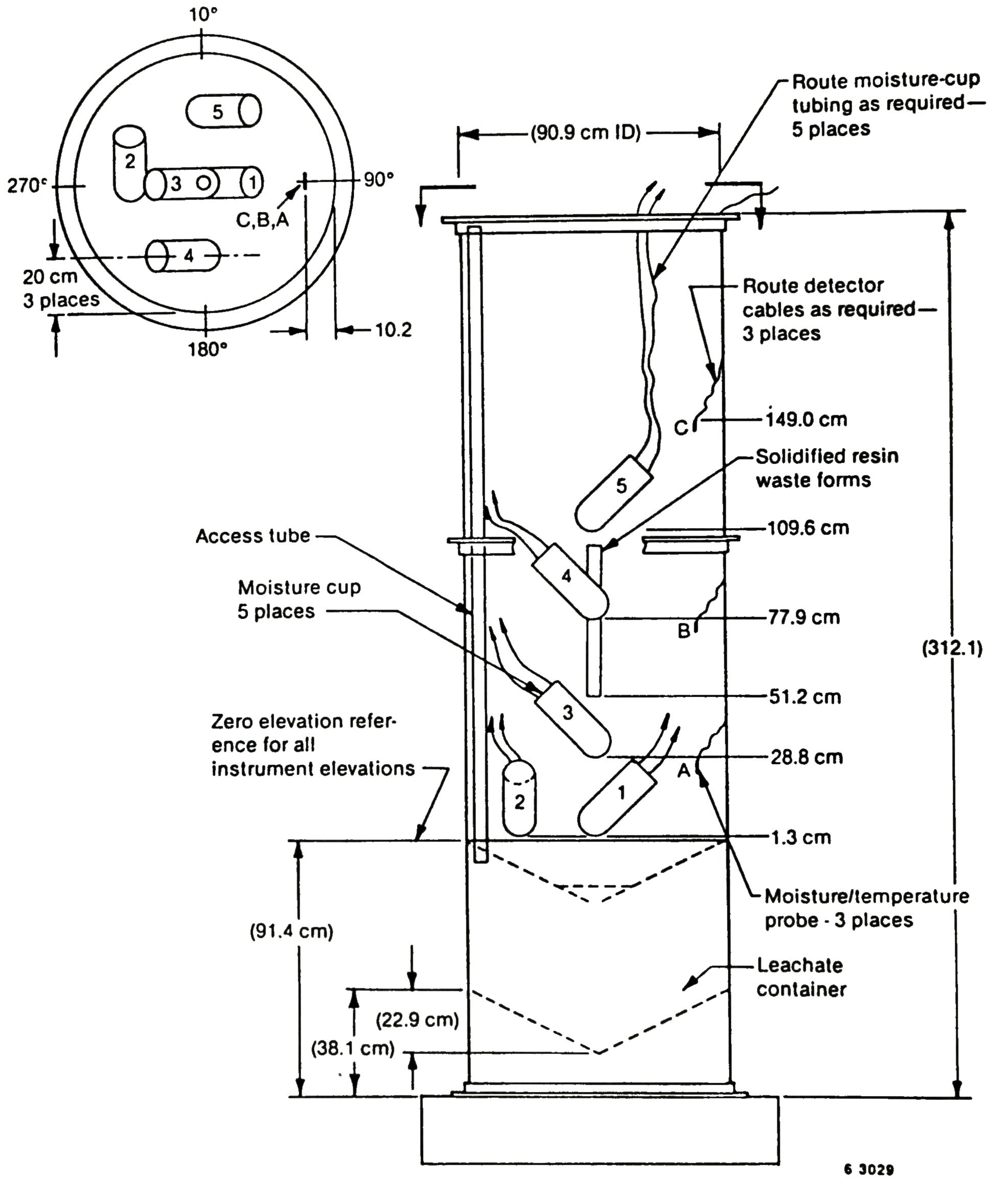

Figure 2. EPICOR-II lysimeter vessel component locations.

Four lysimeters at each field site are filled with soil; the remaining one is a control filled with an inert silica sand.[1] Two different soils were used. One was representative of Midwestern soils; the other was intended to approximate soil found at Barnwell, South Carolina. ANL-E used local indigenous soil which fit the NRC criteria for the Midwestern soil. It is a Morley silt loam, with the surface layer removed. The resulting subsurface soil is a clay loam. Soil at ORNL was not found to be a suitable substitute for Barnwell soil. Therefore, soil acceptable to the NRC was transported to ORNL from the Savannah River Plant adjacent to the Barnwell facility in South Carolina.

Data Retrieval and Analysis

Electrical impulses from the environmental instruments are collected by, processed in, and stored by the DAS for periodic retrieval. The DAS processes input into recognizable data using programmable steps. Output from the soil moisture probes, for example, is processed by a polynomial equation which was derived from laboratory calibration of the probes.[1]

Data output from the DAS is stored on a cassette tape and, after retrieval, is translated to an IBM PC compatible disk file. Hard copy from these files is provided either in a printed format or graphically. The graphic display presents data over an extended time period. The graphic presentation was used for this report.

Water from each lysimeter is drawn from porous cup soil-water samplers and lysimeter leachate collection compartments at least quarterly. These water samples are analyzed routinely for gamma-producing nuclides and were originally scheduled for analysis of the beta-producing nuclide ^{90}Sr on a yearly basis. Water analyses are performed at ANL-E by the Environmental Services Laboratory and at ORNL by the Environmental Radio Analysis Laboratory. Both of these laboratories have a traceable quality assurance program and use accepted analytical procedures for nuclide determination.

RESULTS AND DISCUSSION

The DAS systems functioned well during the first year. There was a period from late August 1985 until November 1985 that the ORNL system was down. Length of the downtime was caused by the necessity of returning the unit to the manufacturer for repairs. No other serious problems have been encountered with these systems. Retrieval, translation, and display of stored data from the two systems have been accomplished with ease.

Weather Data

Wind speed, air temperature, relative humidity, and rainfall, as recorded over a 12-month period by the DAS systems for the ANL-E and ORNL sites, are found on Figures 3 through 10. Rainfall data from the ORNL site appear greatly exaggerated. This trend became apparent during December 1985, and early indications were that the Weather Measure tipping bucket rain gauge supplied with the DAS system was not capable of accurately responding to periods of intense rainfall. In June 1986, this rain gauge was replaced with a Climatronics tipping bucket gauge which is designed for episodic high-intensity rainfall. Data from this gauge appear to be accurate; however, the rainfall data recorded by the DAS contain occasional, erroneously high data points. Corrective measures for determining the source of these spurious data are ongoing. They include monitoring the rain gauge with a separate, single-channel data collection system and testing of the circuits within the DAS system that are responsible for processing rainfall data. This malfunction has not resulted in a loss of rainfall data, since both ANL-E and ORNL have mechanical recording rain gauges in close proximity to their lysimeter sites. Data from those nearby rain gauges (Table 1) were used to calculate the total quantities of precipitation received by each site. Air temperature data from ANL-E (Figure 4) show that there were days of freezing temperatures from mid-November 1985 until mid-March 1986, while there were very few days with air temperatures of 0°C at ORNL (Figure 8).

TABLE 1. YEARLY PRECIPITATION AT ANL-E AND ORNL AS MEASURED BY BACK-UP INSTRUMENTATION--JUNE 1985 THROUGH JULY 1986

Month	Precipitation (cm) ANL	Precipitation (cm) ORNL
July[a]		13.3
August[b]	5.6	23.1
September[c]	6.3	4.3
October	11.6	7.6
November	18.9	10.2
December	1.7	5.3
January	0.6	3.1
February[c]	6.4	10.4
March	7.6	7.1
April[c]	4.0	5.1
May	7.7	7.7
June[c]	11.1	2.6
July	8.6	

a. ORNL lysimeter experiment initiated in July.

b. ANL-E lysimeter experiment initiated in August.

c. Months leachate was retrieved for analyses.

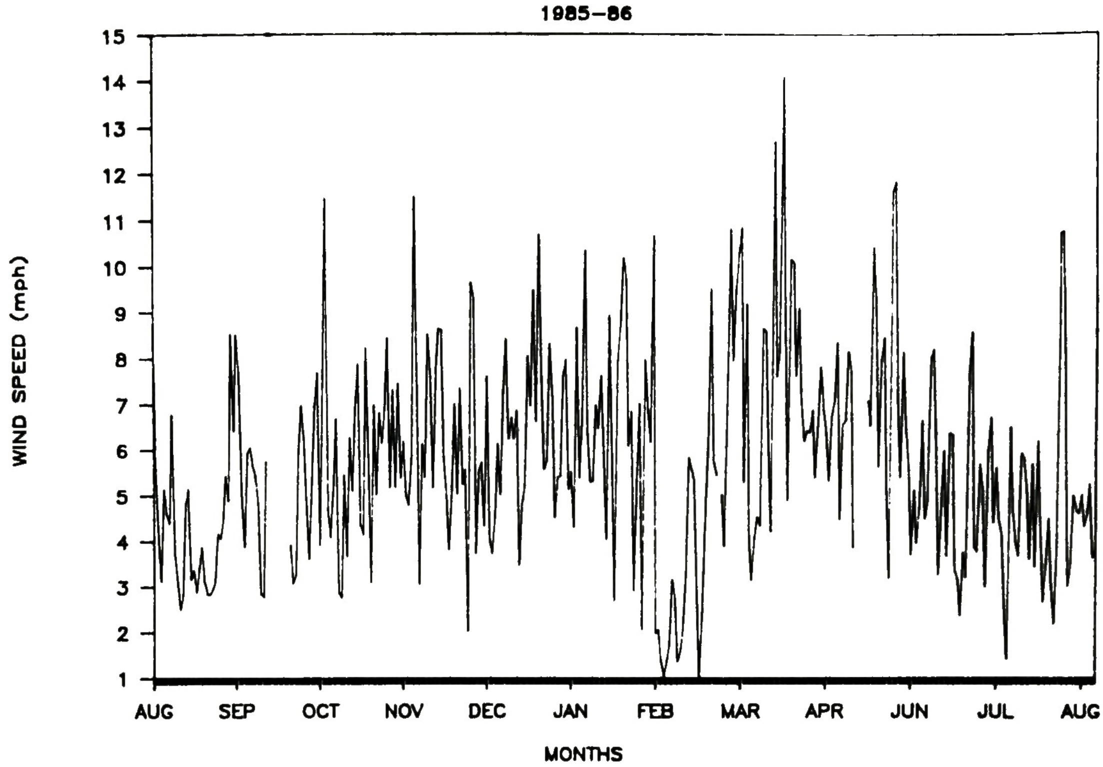

Figure 3. ANL-E weather data--wind speed.

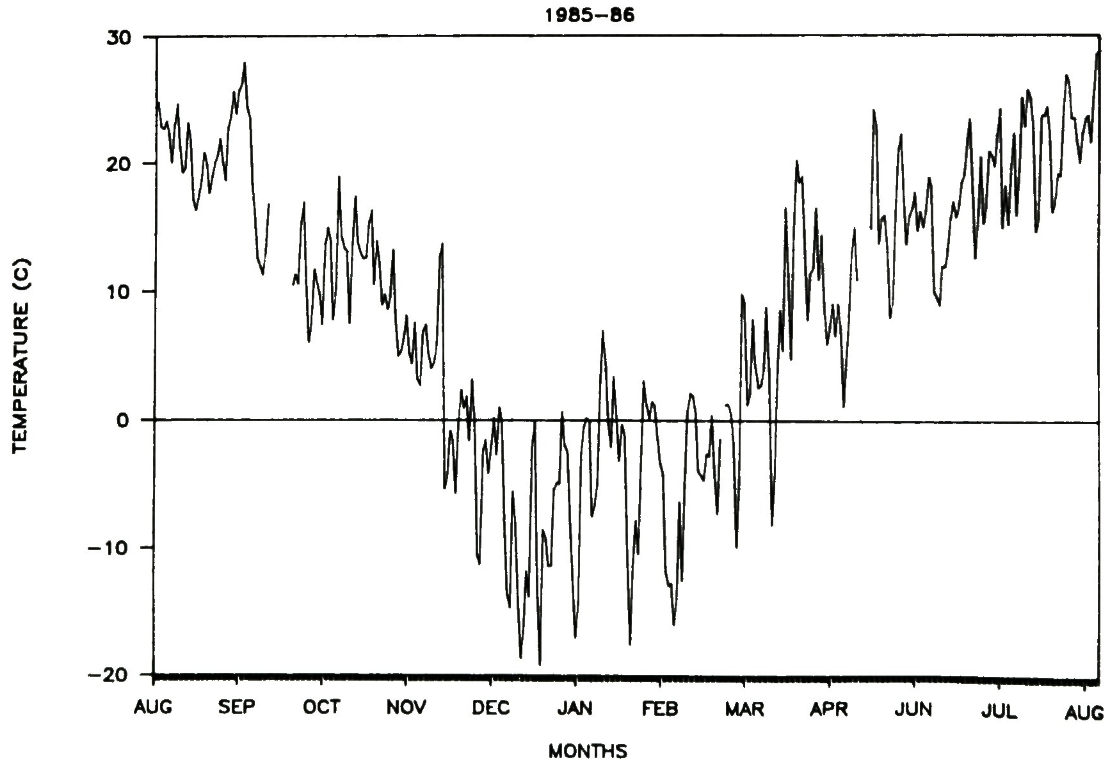

Figure 4. ANL-E weather data--temperature.

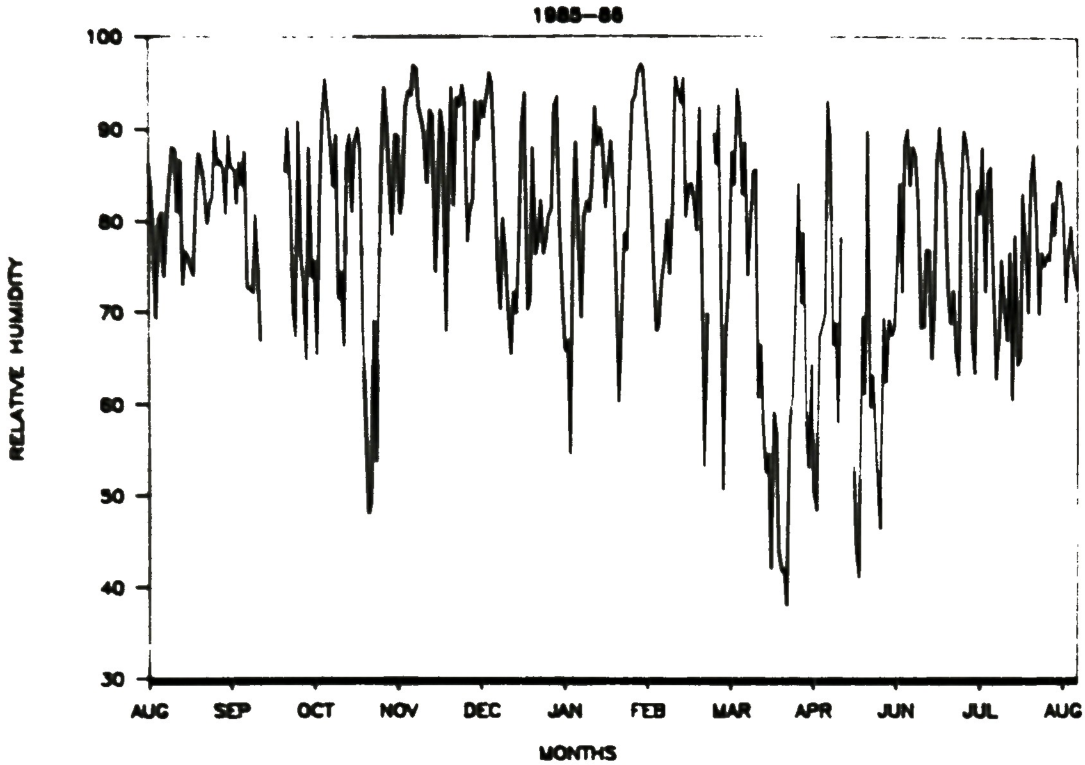

Figure 5. ANL-E weather data--relative humidity.

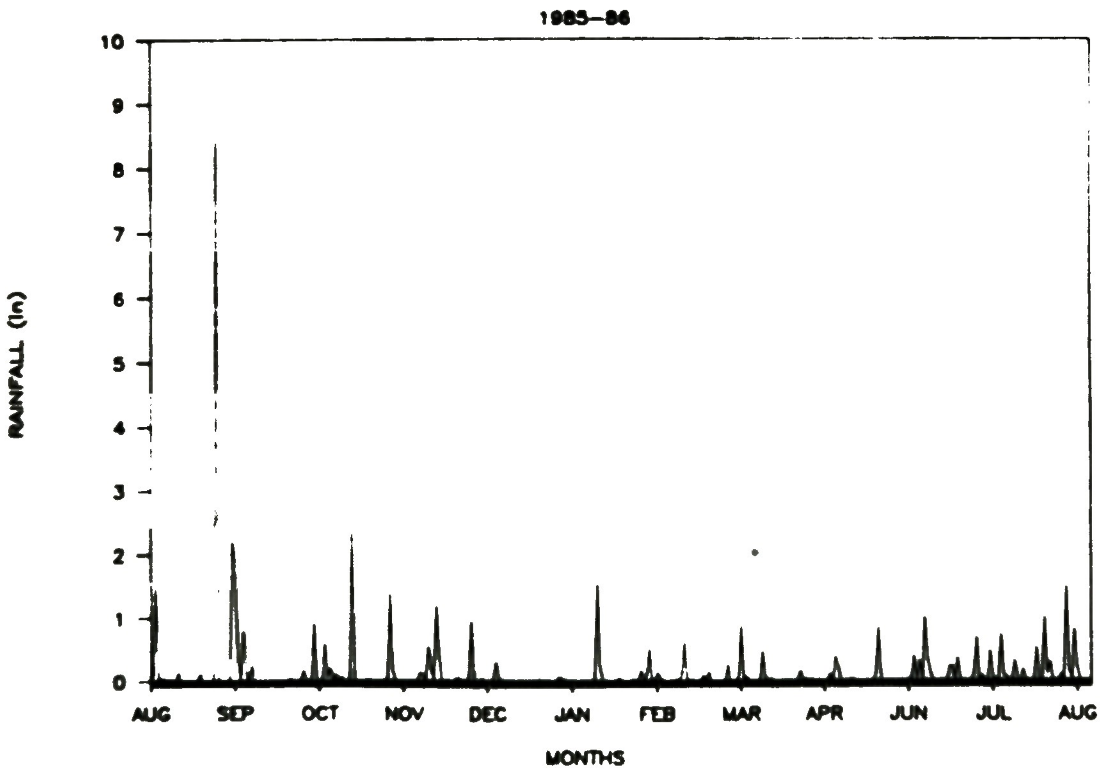

Figure 6. ANL-E weather data--rainfall.

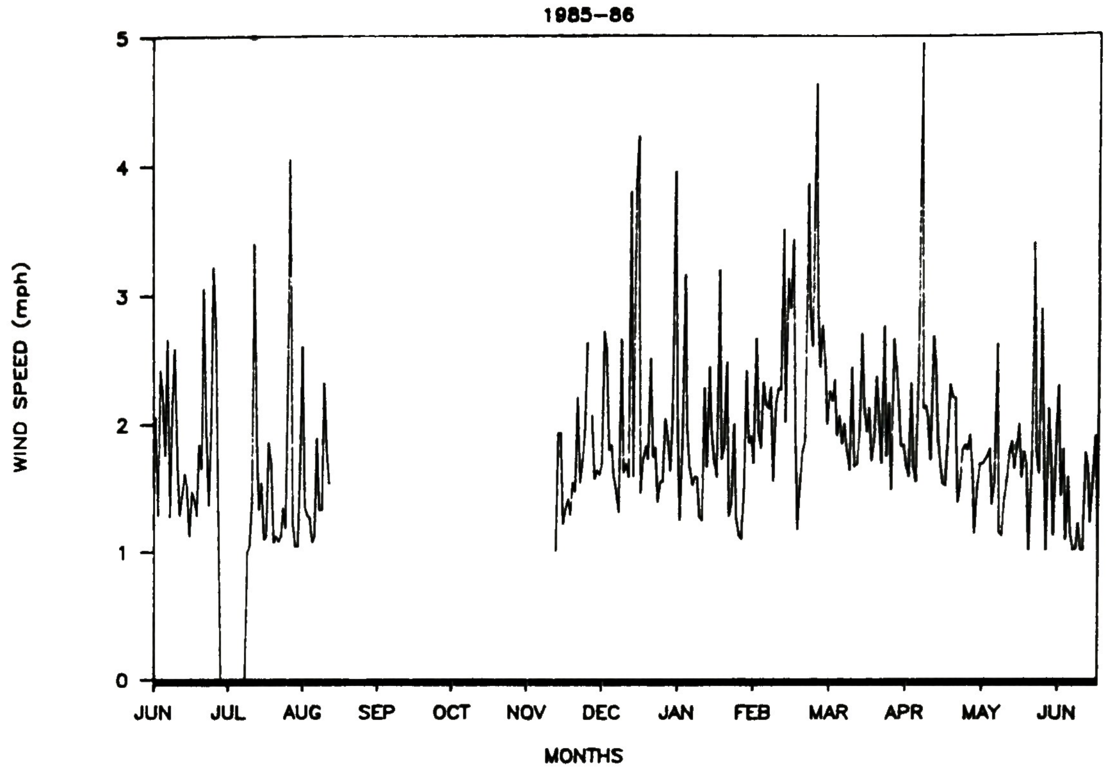

Figure 7. ORNL weather data--wind speed.

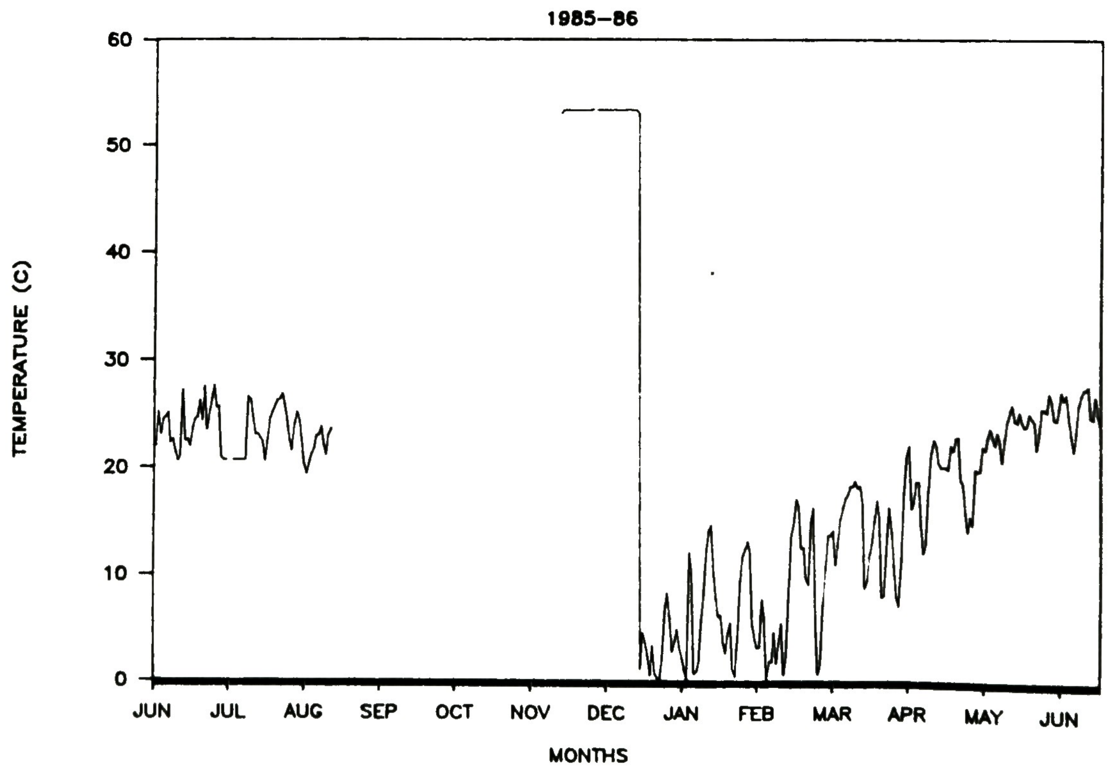

Figure 8. ORNL weather data--temperature.

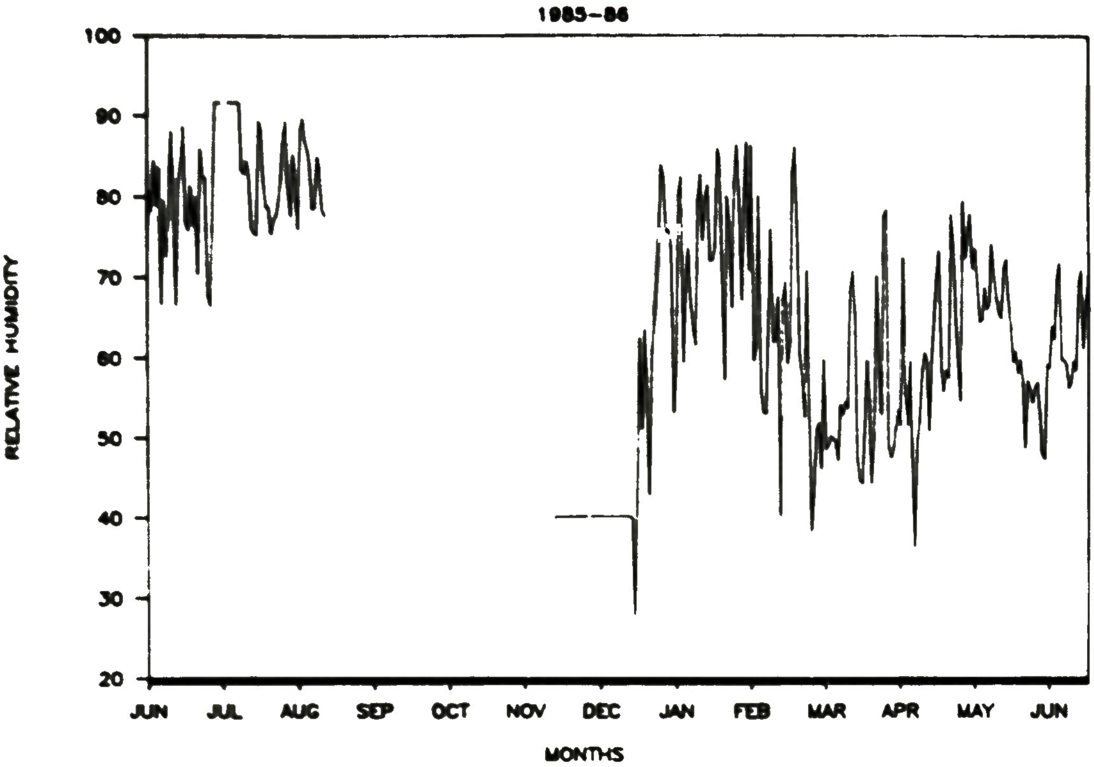

Figure 9. ORNL weather data--relative humidity.

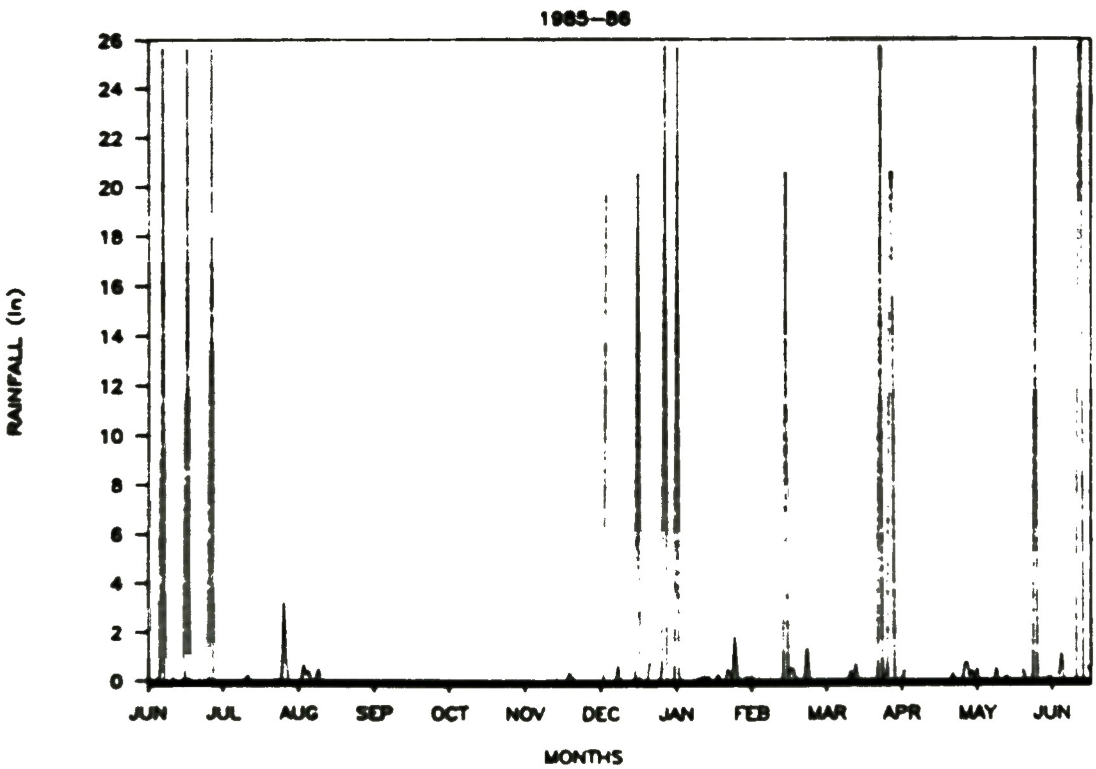

Figure 10. ORNL weather data--rainfall.

Lysimeter Soil Temperature Data

Temperature probes located in all ten lysimeters at the depth of the waste forms (77.9 cm) indicate that at no time were the waste forms exposed to freezing temperatures (Figures 11 through 20). Plots of the soil temperatures further show (as would be expected) that the near-surface soil temperatures (elevation 149.0 cm, 66.7 cm below the soil surface) fluctuate more than the intermediate (elevation 77.9 cm) or bottom (elevation 28.8 cm) soils. It is also noted from these data that the frost line in the soil did not move as deep as the first probe (66.7 cm below the soil surface).

Some abnormally low soil temperature readings were observed from the intermediate and bottom probes in lysimeter ANL-3 in January 1986 and in ANL-4 by June 1986 (Figures 13 and 14). There were no such occurrences with near-surface probes. One possible explanation for the malfunction is related to an average soil subsidence of 30 cm in all ANL-E lysimeters except the sand-filled control. It is hypothesized that subsiding soil may have caused damage to the lead wires connecting the probes to the soil surface. These probes are now being replaced with new ones, and recent data from the replacements shows that they are functioning normally. An example of how closely temperature data from the ANL-E lysimeters tracked each other can be seen in Figures 11, 12, and 15.

The bottom temperature probes in ORNL-3 and -5 have consistently indicated elevated temperature (Figures 18 and 20). Since the abnormal readings began close to the time of lysimeter installation, it is possible that probes or wiring were damaged at that time. In any case, they are to be replaced. All of the other temperature probes at ORNL are functioning, and that includes the ones at the 77.9-cm elevation which are in the proximity of the waste forms. In addition, there is sufficient redundancy in temperature measurement from the functioning probes in the other soil-filled lysimeters to provide adequate data on soil temperatures.

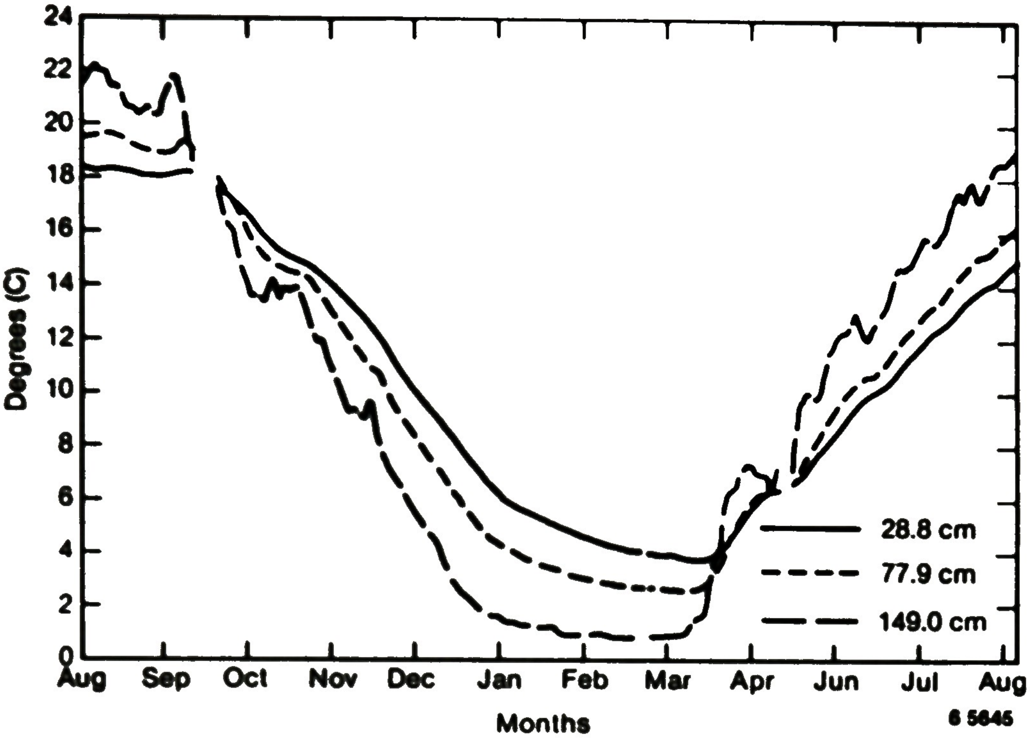

Figure 11. ANL-E Lysimeter 1 soil temperatures.

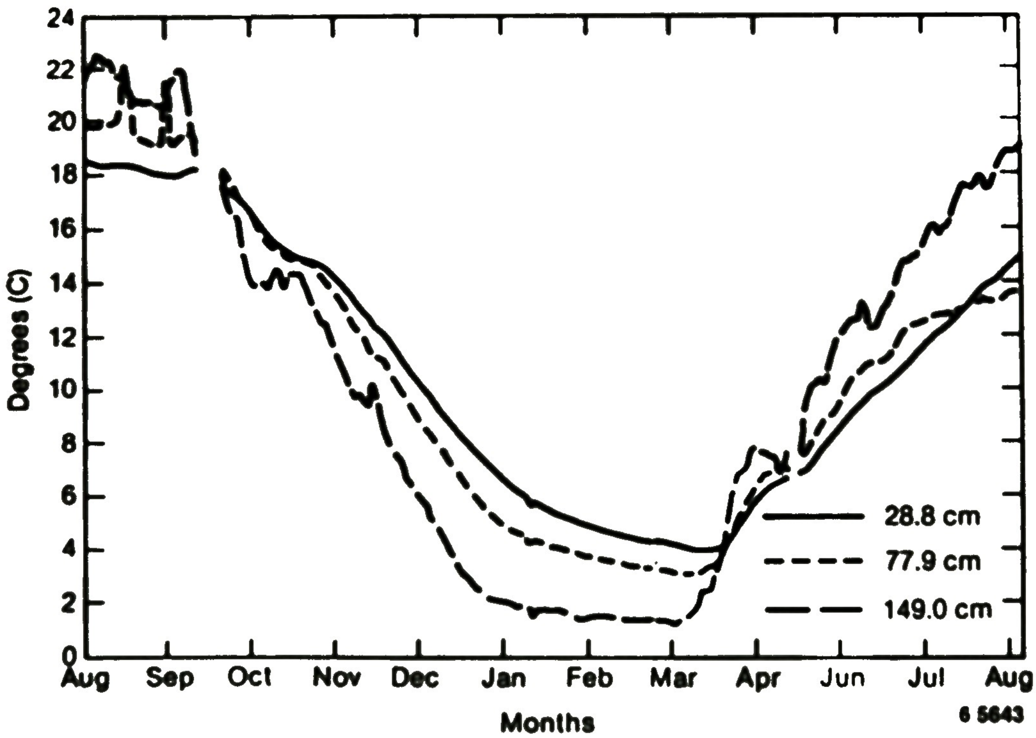

Figure 12. ANL-E Lysimeter 2 soil temperatures.

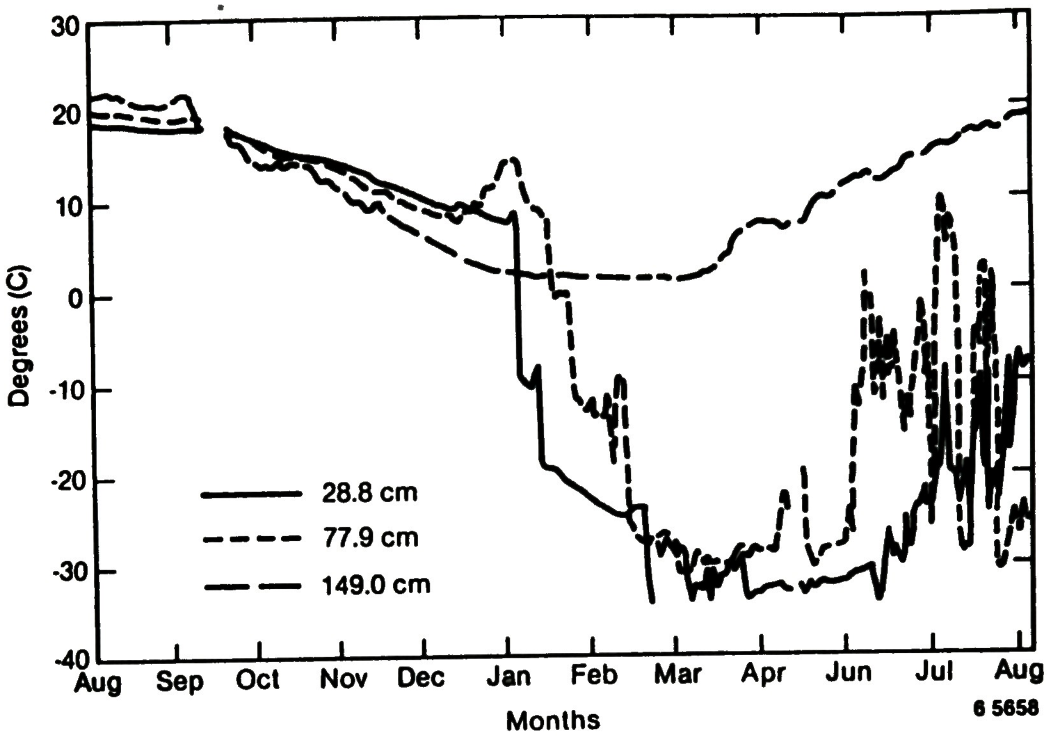

Figure 13. ANL-E Lysimeter 3 soil temperatures.

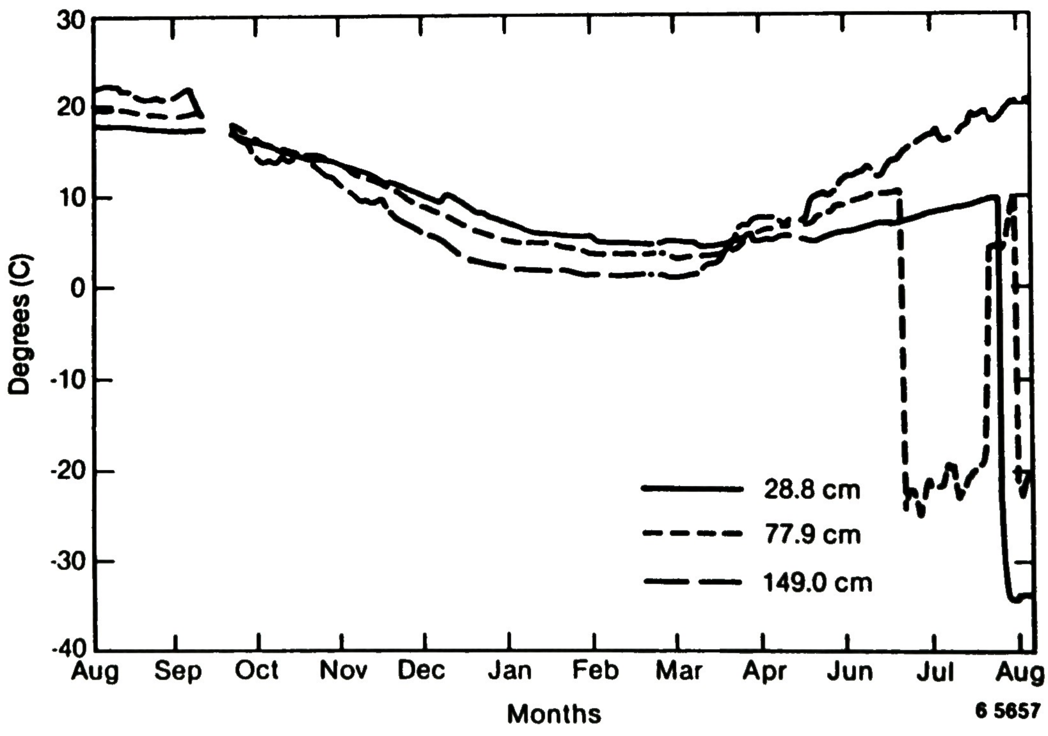

Figure 14. ANL-E Lysimeter 4 soil temperatures.

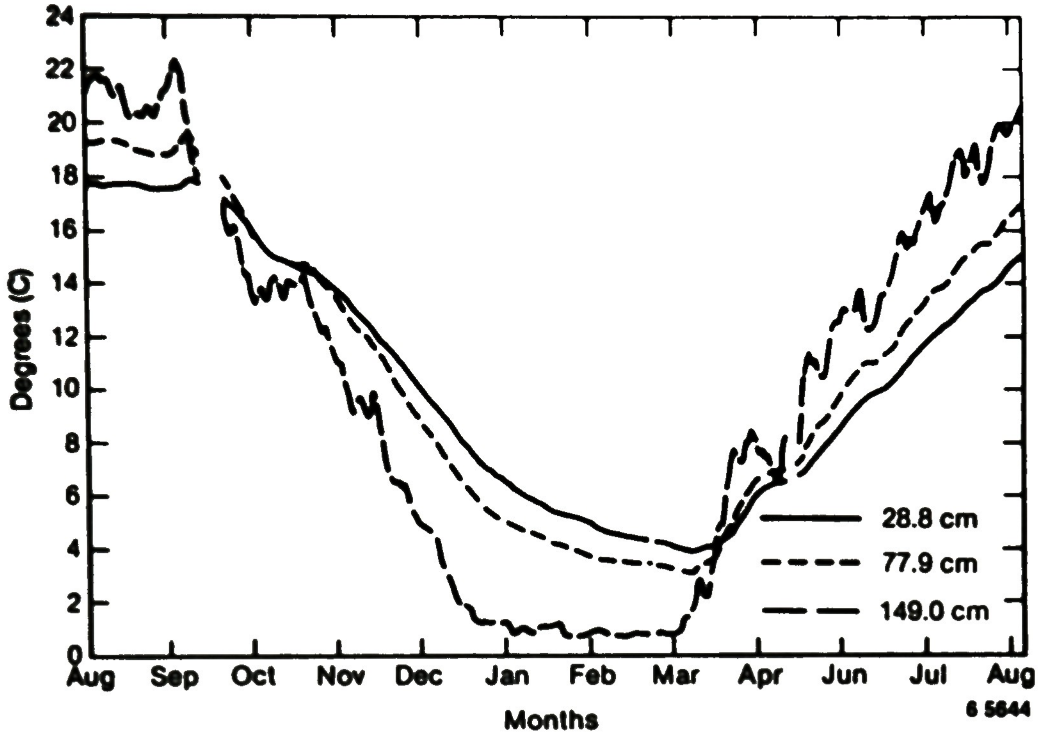

Figure 15. ANL-E Lysimeter 5 soil temperatures.

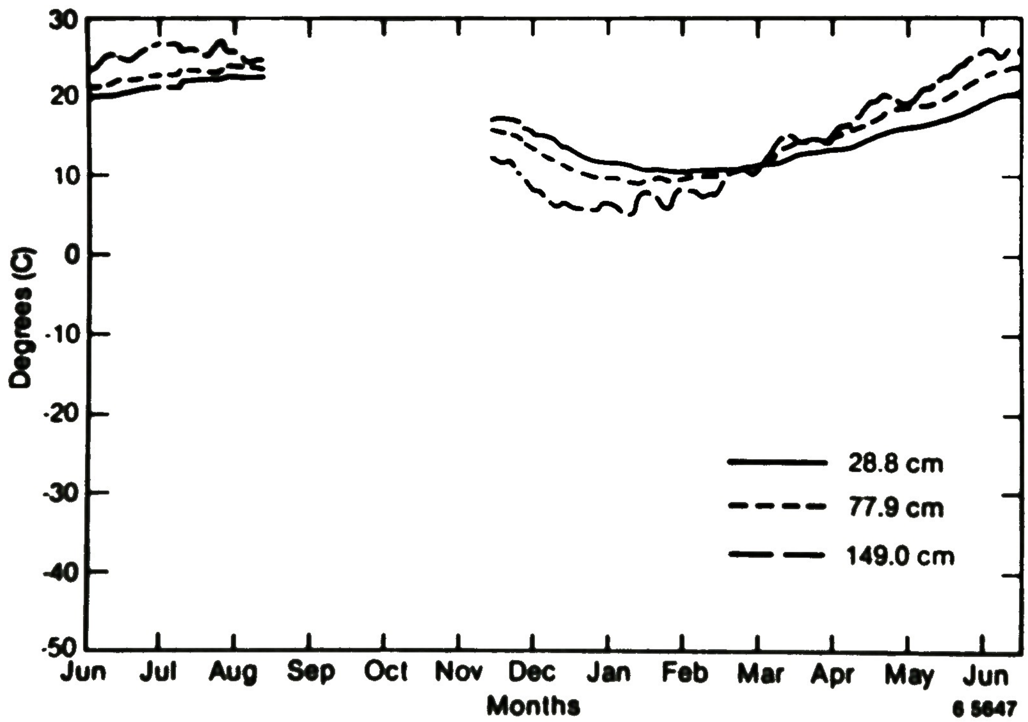

Figure 16. ORNL Lysimeter 1 soil temperatures.

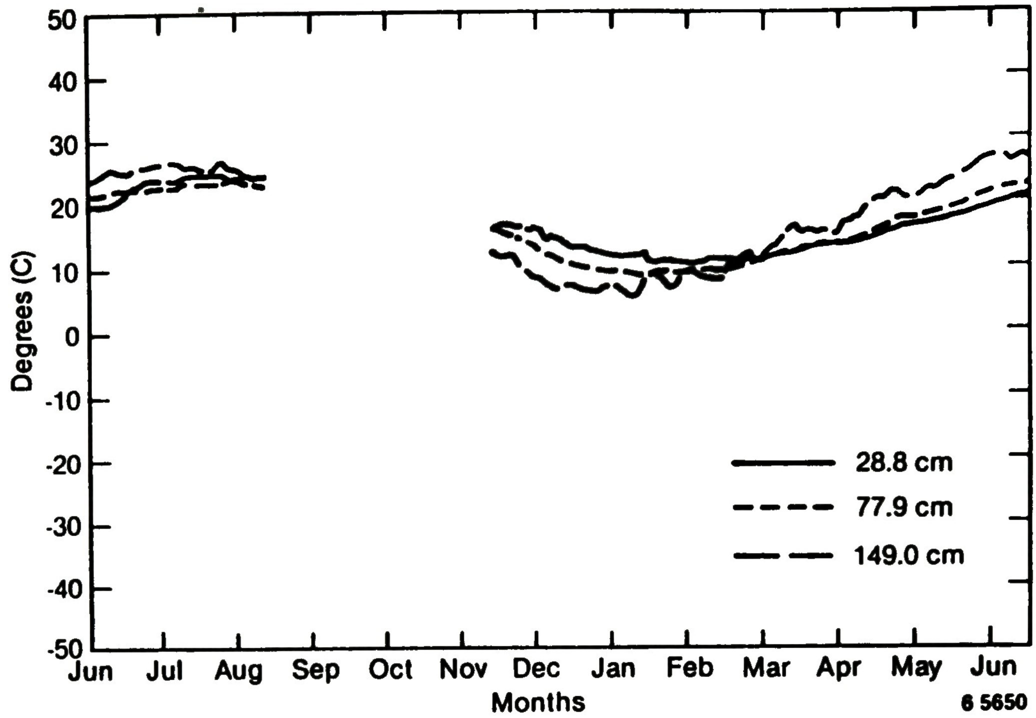

Figure 17. ORNL Lysimeter 2 soil temperatures.

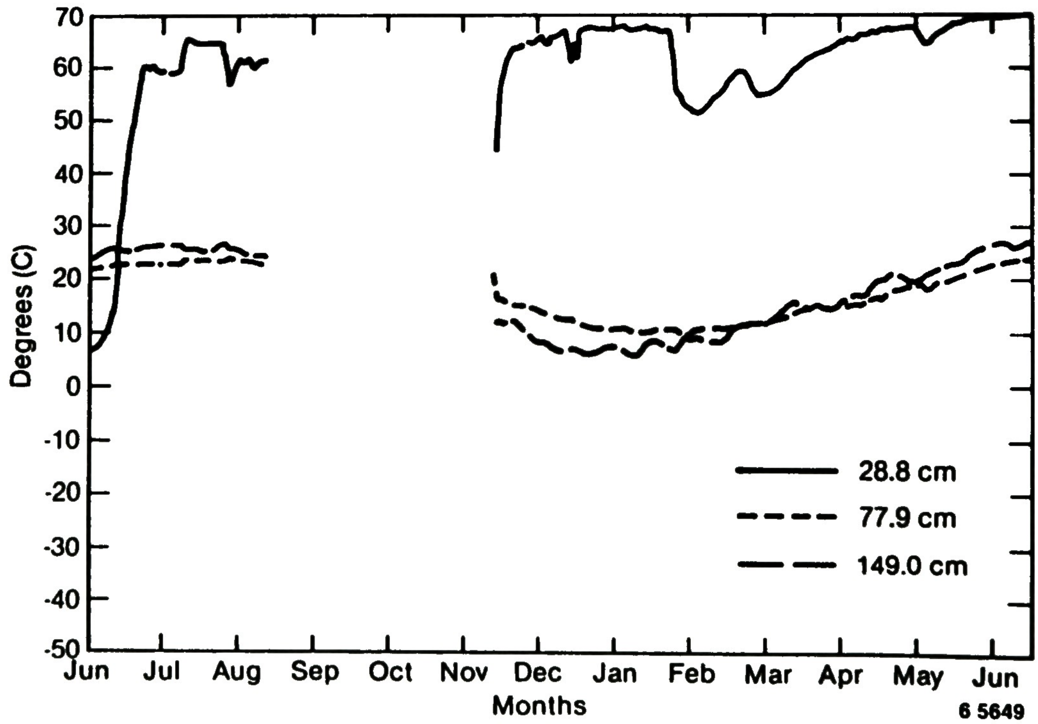

Figure 18. ORNL Lysimeter 3 soil temperatures.

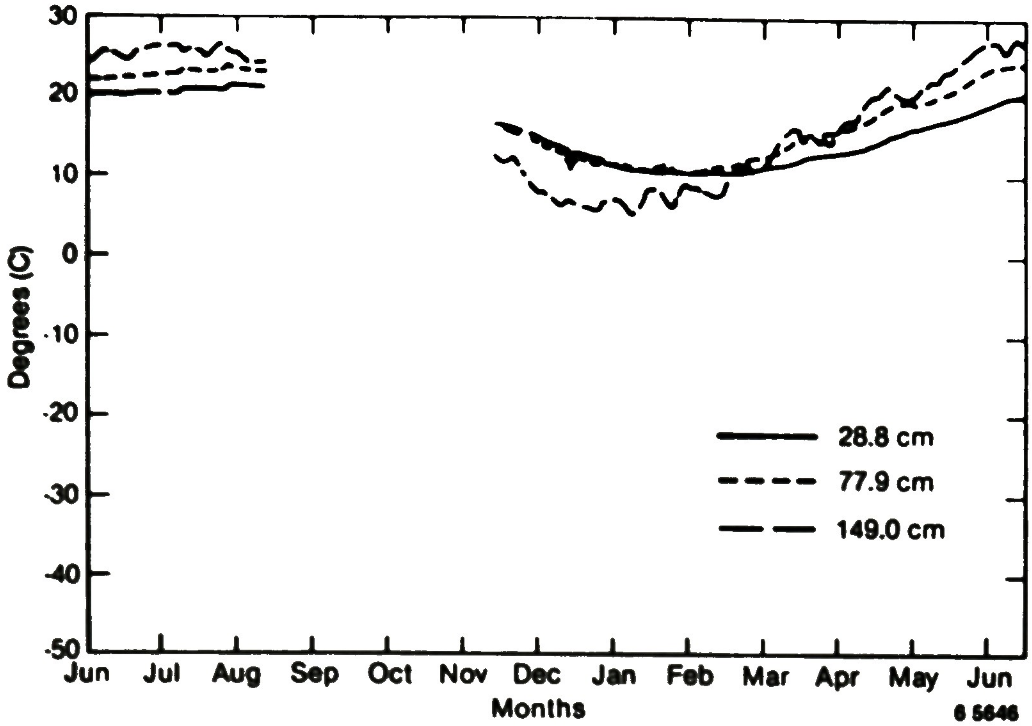

Figure 19. ORNL Lysimeter 4 soil temperatures.

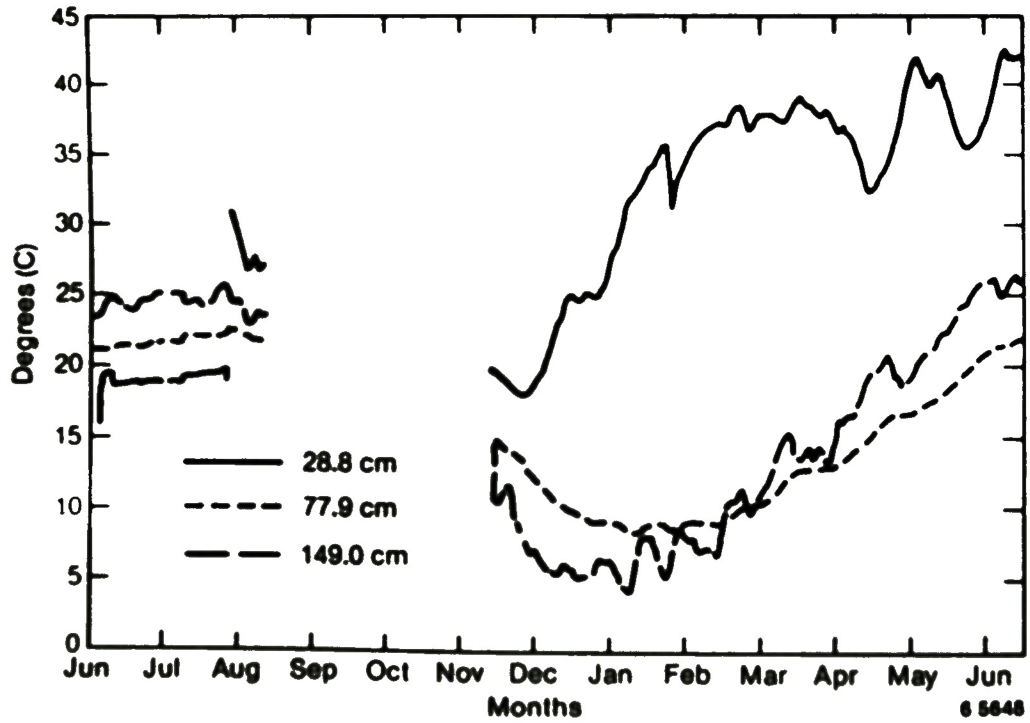

Figure 20. ORNL Lysimeter 5 soil temperatures.

Lysimeter Soil Moisture Data

Moisture probes at the two sites show that two (ORNL) to three (ANL-E) months were required after the lysimeters were filled with soil for the soil to reach saturation (Figures 21 through 30). These data indicate that the waste forms have been exposed to moist soil for nine to ten months. As a precaution, the accuracy of the probes in the soil-filled lysimeters was determined by comparing their data against the gravimetric water content of soil cores retrieved from all four ORNL lysimeters and one ANL-E lysimeter. It is apparent from that data (Tables 2 and 3) that the probes are over-estimating the soil moisture content. Corrective action is ongoing and consists of recalculation of the polynomial equation which transforms probe input into percent moisture. Data for recalculation of the equation are coming from laboratory recalibration of several soil moisture probes using lysimeter soils. If the laboratory calibration does not produce satisfactory results, the probe manufacturer suggests that field calibration be used. This would be accomplished by measuring soils near the surface probes over an extended period of time. The moisture content of soil samples through one or two wetting and drying cycles would be determined gravimetrically. A polynomial would then be calculated using those data.

During a 12-month period, the ANL-E site had 93.5 cm of precipitation. ORNL received 99.0 cm, well below the normal of 134 cm. Using these values and the area of exposed lysimeter (6489.5 cm^2), it is calculated that the ANL-E and ORNL lysimeters received 607 L and 643 L of water, respectively. Total water retrieved from the leachate collectors of each lysimeter is shown in Table 4. On the average, the collectors of the soil-filled ANL-E lysimeters contained 128.7 ± 22.6 L; those at ORNL, 441.7 ± 20.9 L. The collectors of the sand-filled lysimeters at ANL-E and ORNL contained 337.9 L and 528.0 L, respectively.

The lysimeters at each site received comparable volumes of water, though those quantities did not move through the lysimeters at each site in equal amounts due to differences in soil texture and weather conditions.

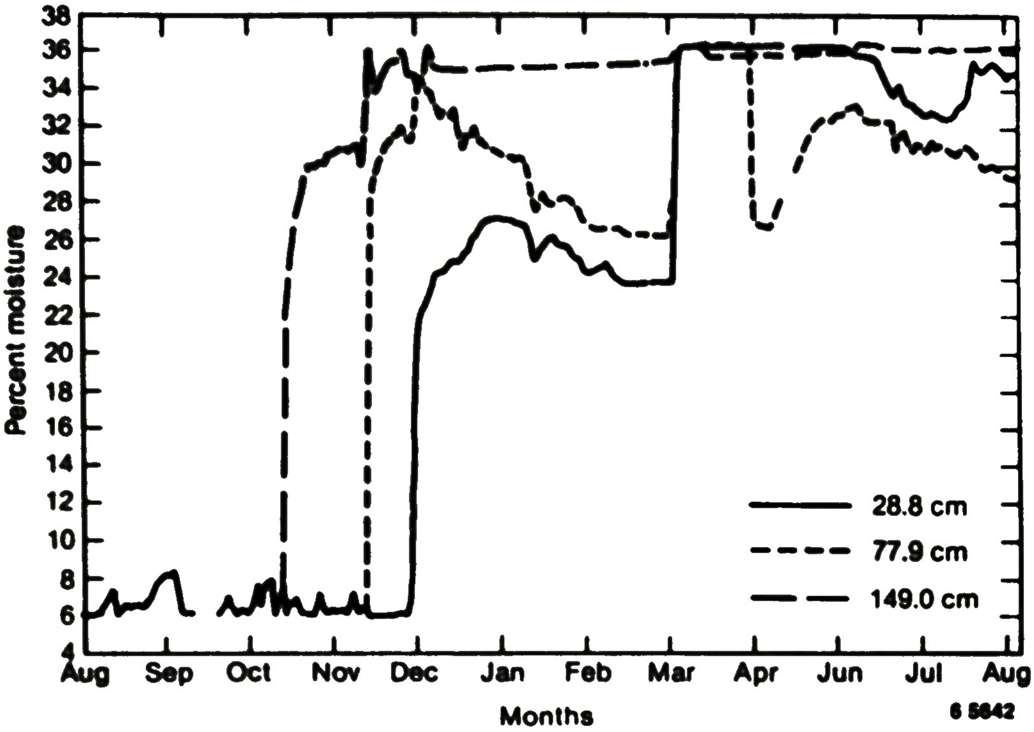

Figure 21. ANL-E Lysimeter 1 soil moistures.

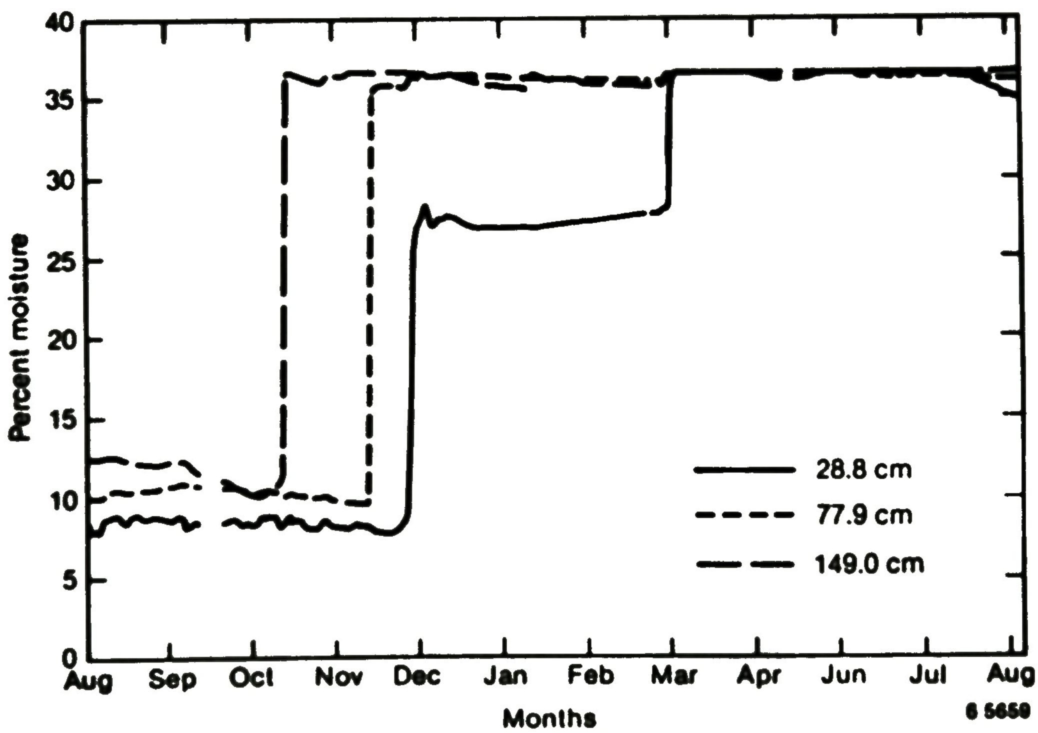

Figure 22. ANL-E Lysimeter 2 soil moistures.

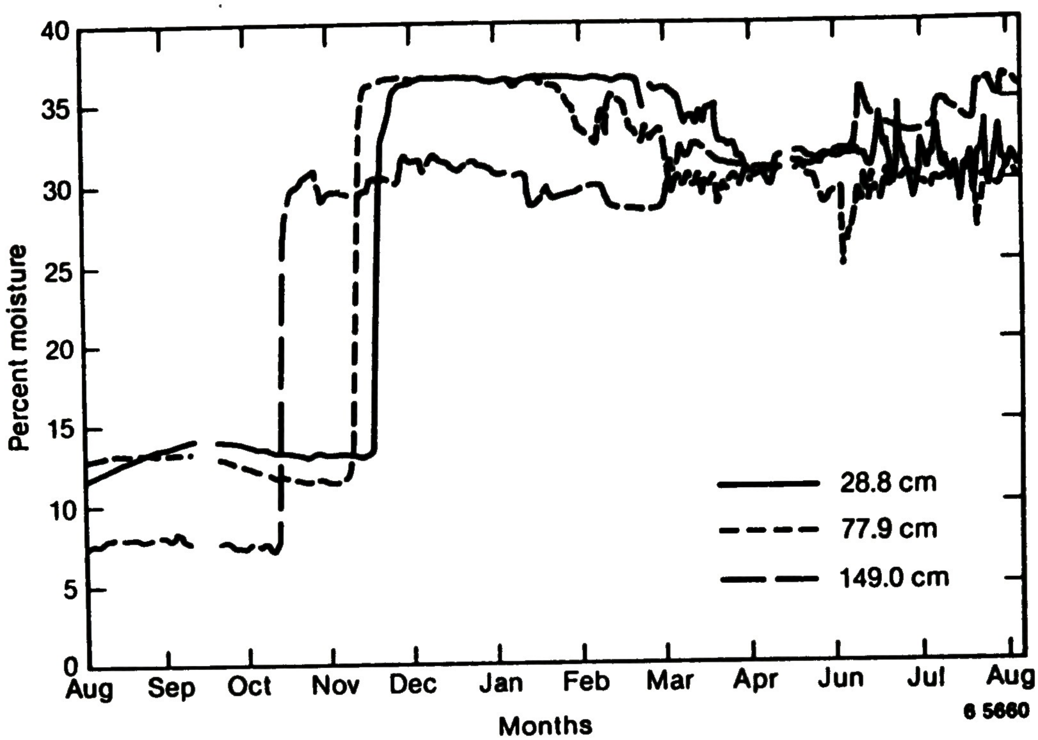

Figure 23. ANL-E Lysimeter 3 soil moistures.

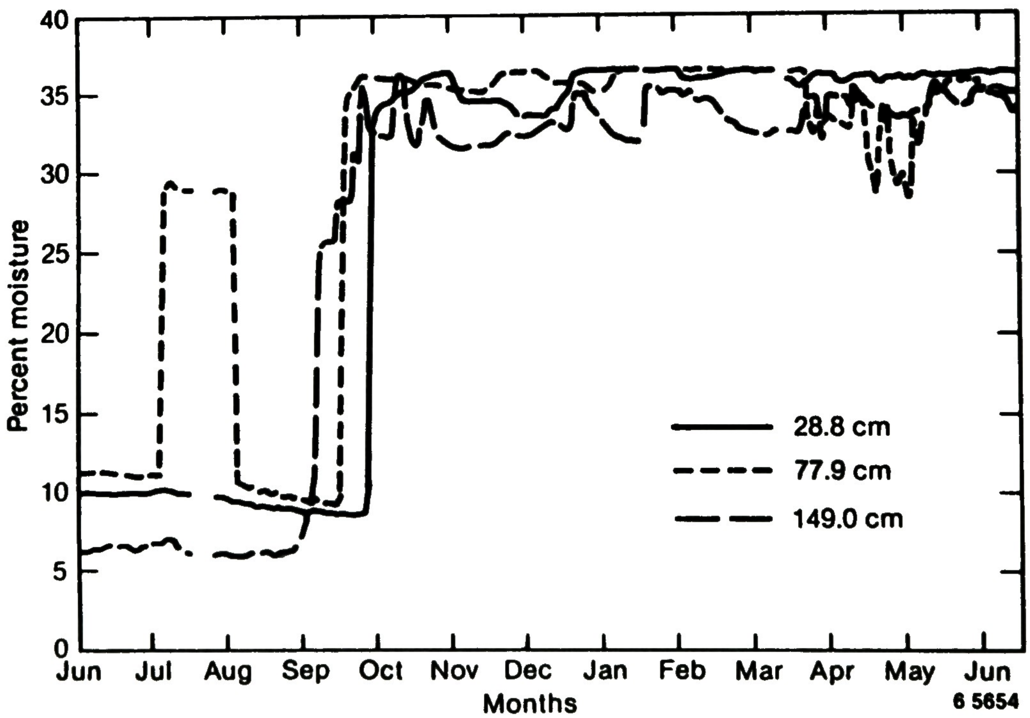

Figure 24. ANL-E Lysimeter 4 soil moistures.

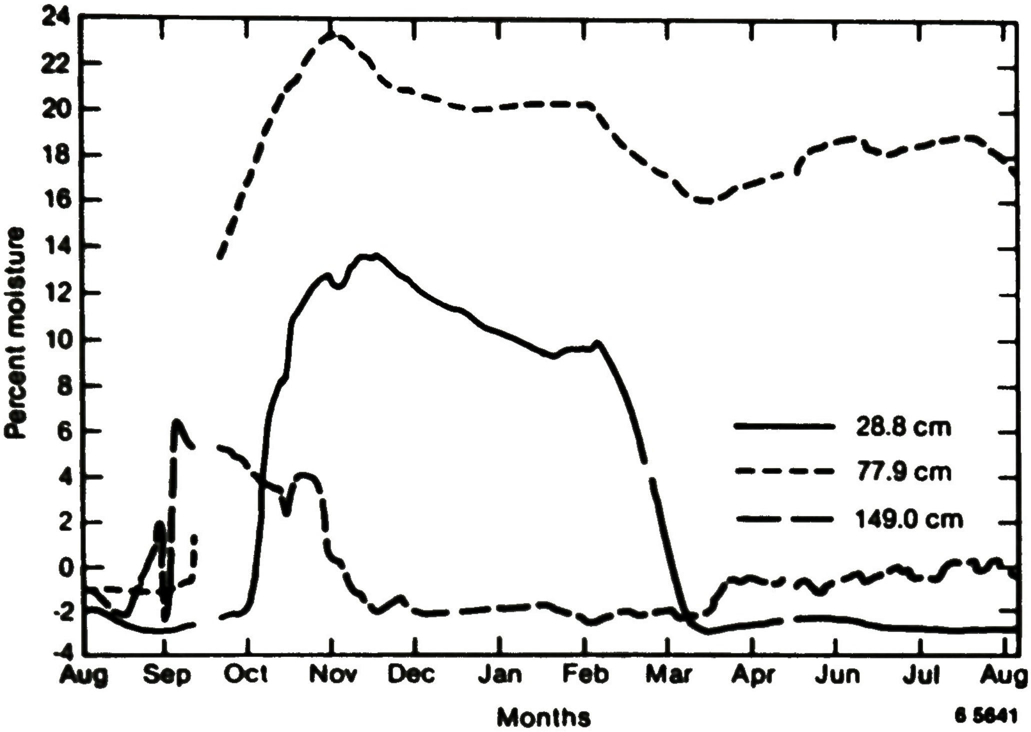

Figure 25. ANL-E Lysimeter 5 soil moistures.

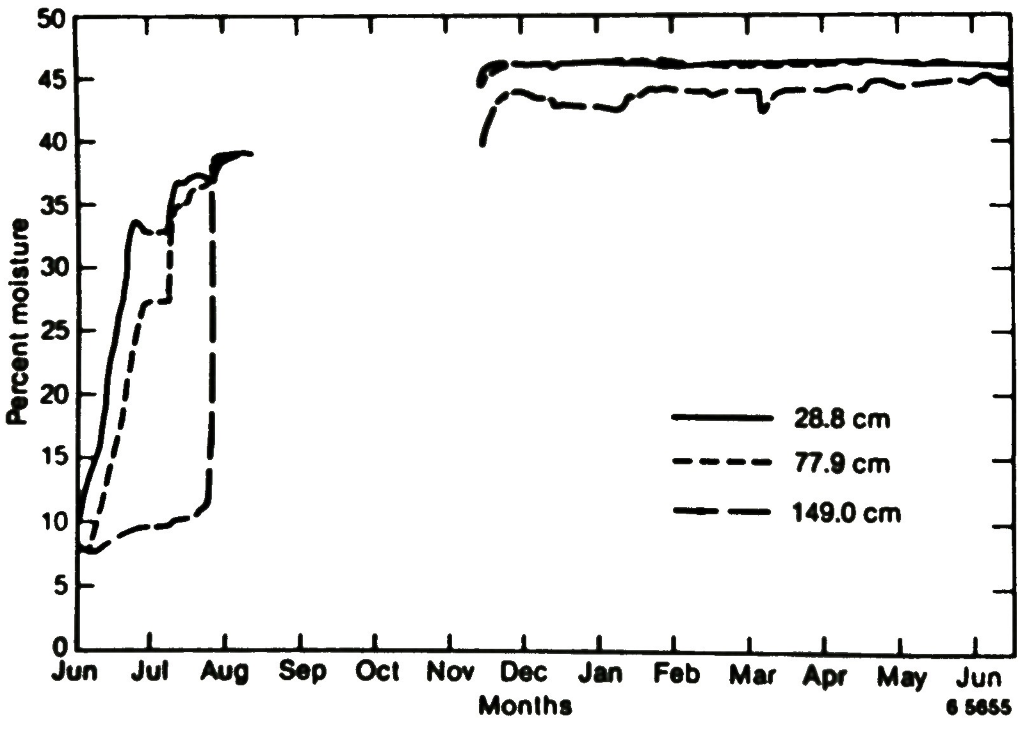

Figure 26. ORNL Lysimeter 1 soil moistures.

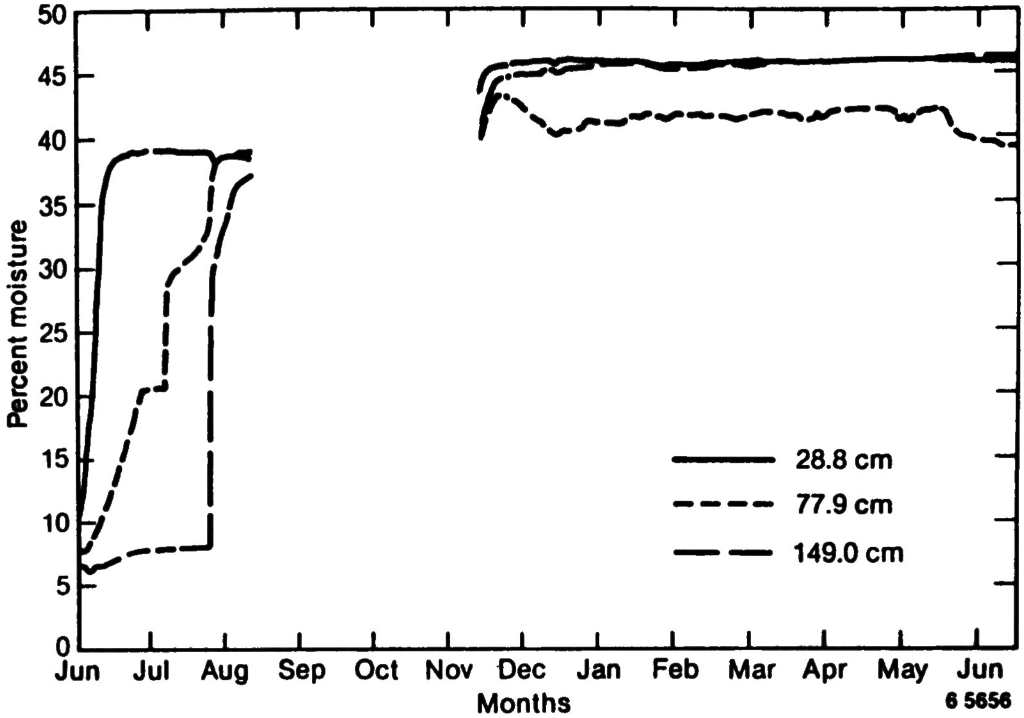

Figure 27. ORNL Lysimeter 2 soil moistures.

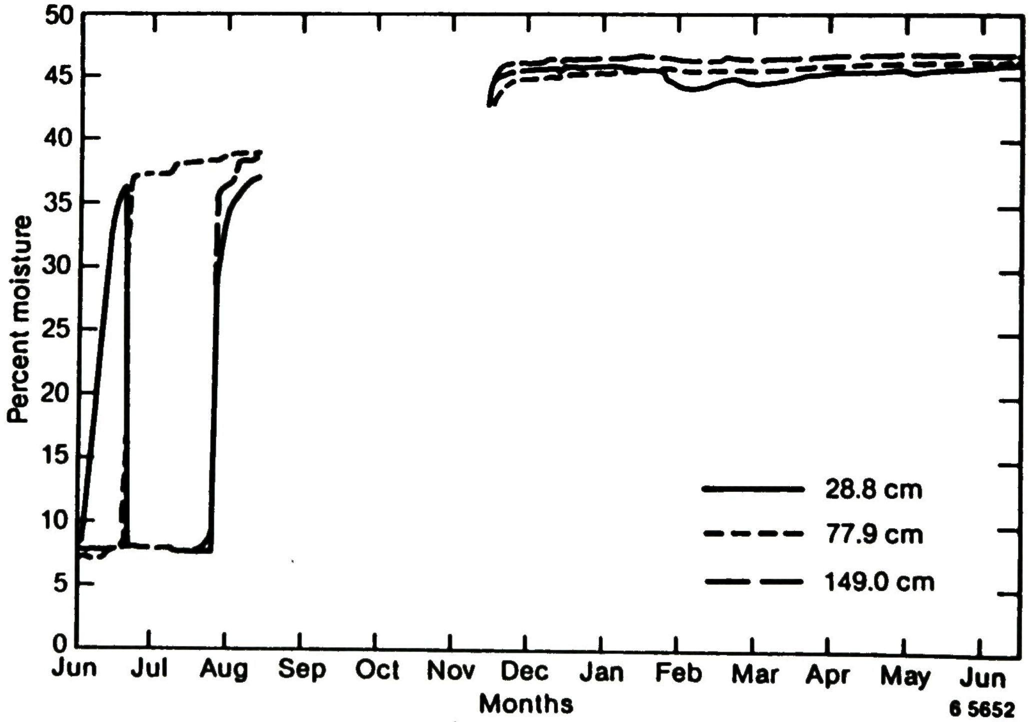

Figure 28. ORNL Lysimeter 3 soil moistures.

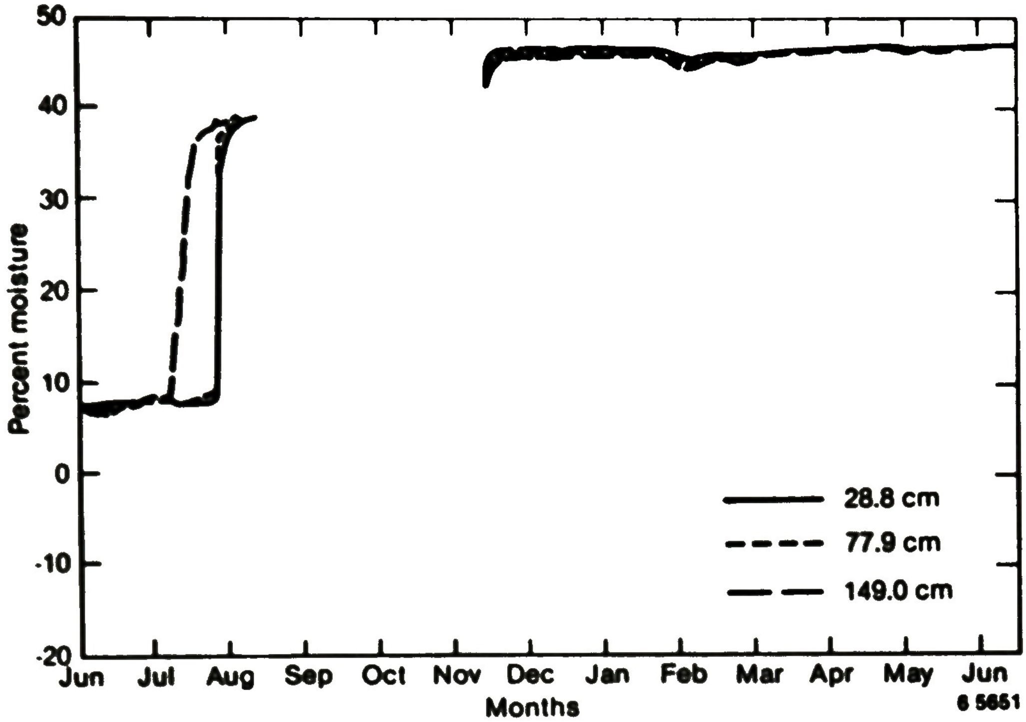

Figure 29. ORNL Lysimeter 4 soil moistures.

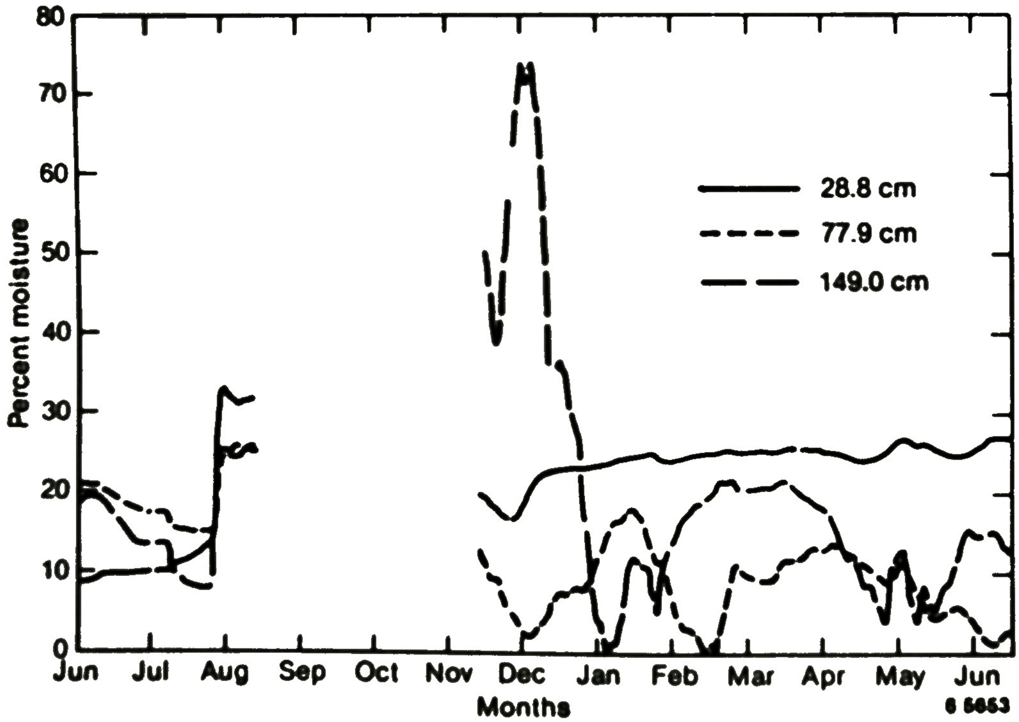

Figure 30. ORNL Lysimeter 5 soil moistures.

TABLE 2. MOISTURE PROFILE OF ANL-E LYSIMETER 3 BASED ON GRAVIMETRIC MEASUREMENT OF WATER CONTENT

Depth (cm)	Wet Weight (g)	Dry Weight (g)	Moisture (Dry Wt) (%)
0-20	15.76	13.42	17.4
20-41	21.84	18.21	19.9
41-61	22.87	18.80	21.0
61-81	28.32	23.04	22.9
81-102	18.70	15.24	22.7
102-122	14.04	11.37	23.4
122-142	17.03	13.83	23.1
142-162	19.08	15.54	22.7
162-183	19.18	15.42	24.3

TABLE 3. MOISTURE PROFILE OF ORNL LYSIMETERS 1 THROUGH 4 BASED ON GRAVIMETRIC MEASUREMENT OF WATER CONTENT

Lysimeter	Depth (cm)	Wet Weight (g)	Dry Weight (g)	Moisture (Dry Wt) (%)
1	0-36	105.58	92.85	12.5
1	36-71	97.84	85.66	14.2
1	71-107	77.82	67.63	15.1
2	0-36	101.07	88.90	13.7
2	36-71	72.12	63.18	14.1
2	71-107	92.98	81.35	14.3
3	0-36	102.95	91.31	12.7
3	36-71	72.33	63.12	14.6
3	71-107	72.32	62.78	15.2
4	0-36	92.49	82.00	12.8
4	36-71	87.14	76.14	13.9
4	71-107	71.13	62.02	14.7

TABLE 4. TOTAL QUANTITIES OF LEACHATE RETRIEVED FROM LYSIMETERS DURING A 12-MONTH PERIOD

	Quantity of Water (L)				
	Lysimeter Number				
Site	1	2	3	4	5
ORNL	415	438	449	465	528
ANL-E	113	132	160	112	338

Soil used at ANL-E is heavier (contains more fine material such as silts and swelling clay) than the soil used at ORNL.[1] Therefore, infiltration and percolation of water through the ANL-E soil would be expected to be significantly reduced in comparison to ORNL soil. The effect of weather is especially apparent when comparing the sand-filled control lysimeters at the two sites. At ANL-E, 55% of the volume of precipitation passed through the lysimeter versus 82% for ORNL. During the year, 42% of the ANL-E precipitation came during the months of November through March when the average air temperature was below 0°C. This precipitation then was in the form of freezing rain or snow which would not penetrate the frozen soil surface and could have been blown off (in the case of snow) or lost due to sublimation. Other factors such as generally gustier winds and lower humidity at ANL-E indicate that evaporation of water from the ANL-E lysimeters would be decidedly higher than at ORNL. (Wind speed and relative humidity for ANL-E and ORNL are shown in Figures 3, 5, 7, and 9.)

Based on the amount of water retrieved from the lysimeters, the ANL-E soil lysimeters had 0.18 pore volumes of water pass through them while the similar ORNL lysimeters had 0.62 pore volumes. Pore volume quantities for the control lysimeters were 0.57 for ANL-E and 0.94 at ORNL. Theoretically then, 18% of the water held in the soil pore space of the ANL-E lysimeters was replaced during the year, while 62% was replaced in the ORNL soil lysimeters and 57% and 94% was replaced in the ANL-E and ORNL control lysimeters, respectively. Therefore, if nuclides were in the water surrounding the waste forms, the greatest opportunity for detection would be found in water from the ORNL site. (This is based on two assumptions: (a) the nuclide is water soluble; and (b) the soil column does not interfere with nuclide movement.)

Radionuclide Analysis

Water samples were collected from the leachate collectors and moisture cup 3 from each lysimeter on four occasions during the year (see Table 1). The first two times, water samples were analyzed only for gamma-producing nuclides. The latest water samples, taken at both sites in April and

June 1986, were analyzed for both gamma-producing nuclides and the beta-producing nuclide ^{90}Sr. Results of those analyses are given in Tables 5 through 8.

During June, in addition to obtaining water samples from leachate collectors and moisture cup 3, water samples were taken from moisture cup 5 (the one nearest the soil surface) of each soil lysimeter. Those samples were then combined for use as a composite sample. Because moisture cup 5 is located above the waste forms, the composite water sample serves as a control to detect nuclides which might originate from sources other than the waste forms.

Gamma-producing nuclides were not found in the first two samplings. However, in April, ^{60}Co was discovered in water samples from the moisture cup of ANL 3; ^{137}Cs was found in the leachate of ANL 5 (the inert lysimeter); and ^{125}Sb was found in the moisture cup of ORNL 5 (also an inert lysimeter). In addition, ^{90}Sr was found in significant quantities in the moisture cups of ANL 4 and 5 and in the leachate of all ANL-E and ORNL lysimeters during the April sampling. The concentration of ^{90}Sr in the ORNL lysimeter leachate was almost two orders of magnitude higher than the ANL-E samples.

Analysis of June water samples showed that ^{60}Co still persisted in the moisture cup of ANL 3, with a substantial increase in ^{125}Sb in ORNL 5-3. The origin of ^{125}Sb is not known but is assumed to be the waste forms. Original evaluation of radionuclide content of the prefilters from which this resin was taken identified ^{125}Sb in quantities of 0.1% of total nuclide content, although resin analysis has not found any. Cobalt-60 was also found for the first time in ORNL 5-3. Also, ^{90}Sr was detected in moisture cups at ORNL (ORNL 3-3 and 5-3) and in two additional cups at ANL-E (ANL 3-3 and 5-1), while there was none detected in this sampling of ANL 4-3. The concentration of ^{90}Sr in ANL 5-3 was more than double that found in the April sample. ANL 5-1 is the moisture cup located directly below 5-3, and the water from this moisture cup was analyzed for the first time in June in an attempt to detect movement of ^{90}Sr through the silica sand profile. The concentration of ^{90}Sr in ANL 5-1 was almost

TABLE 5. RESULTS OF GAMMA-RAY AND STRONTIUM ANALYSES OF ANL-E SOIL MOISTURE AND LEACHATE SAMPLES--APRIL 1986

	Concentration[a] (pCi/L)		
Sample Identification	Cobalt-60	Cesium-137	Strontium-90
Lys 1-3[b]	<5	<5	1.0 ± 1.8
Lys 2-3	<5	<5	1.1 ± 1.2
Lys 3-3	11 ± 7	<5	1.1 ± 1.0
Lys 4-3	<5	<5	2.7 ± 1.8
Lys 5-3	<5	<5	55.6 ± 3.1
Lys 1[c]	<5	<5	0.5 ± 0.3
Lys 2	<5	<5	0.5 ± 0.2
Lys 3	<5	<5	0.4 ± 0.1
Lys 4	<5	<5	0.6 ± 0.3
Lys 5	<5	5.4 ± 1.1	1.0 ± 0.4

a. Concentration ± 2 sigma.

b. Moisture cup identity number.

c. Leachate collector identity number.

TABLE 6. RESULTS OF GAMMA-RAY AND STRONTIUM ANALYSES OF ORNL SOIL MOISTURE AND LEACHATE SAMPLES--APRIL 1986

Sample Identification	Concentration[a] (pCi/L)			
	Cobalt-60	Cesium-137	Antimony-125	Strontium-90
Lys 1-3[b]	<16	<14	<27	2.7 ± 4.9
Lys 2-3	<16	<14	<27	7.0 ± 13.5
Lys 3-3	<16	<11	<27	5.9 ± 12.9
Lys 4-3	<14	<11	<27	<11
Lys 5-3	<14	<14	351 ± 5	6.2 ± 18.1
Lys 1[c]	<2	<1	<3	62.2 ± 8.1
Lys 2	<1	<1	<3	27.0 ± 5.4
Lys 3	<3	<2	<5	4.9 ± 2.7
Lys 4	<2	<2	<3	54.1 ± 8.1
Lys 5	<2	<1	<5	45.9 ± 8.1

a. Concentration ± 2 sigma.

b. Moisture cup identity number.

c. Leachate collector identity number.

TABLE 7. RESULTS OF GAMMA-RAY ANALYSIS OF ANL-E SOIL MOISTURE AND LEACHATE SAMPLES--JUNE 1986

Sample Identification	Concentrations[a] (pCi/L)		
	Cobalt-60	Cesium-137	Strontium-90
Composite[b]	<5	<5	<1
Lys 1-1[c]	<5	<5	<1
Lys 1-3	<5	<5	<1
Lys 2-3	<5	<5	<1
Lys 3-3	13 ± 7	<5	11.3 ± 1.4
Lys 4-3	<5	<5	<1
Lys 5-1	<5	<5	349.6 ± 11.3
Lys 5-3	<5	<5	127.6 ± 6.7
Lys 1[d]	<1	<1	<1
Lys 2	<1	<1	<1
Lys 3	<1	<1	<1
Lys 4	<1	<1	<1
Lys 5	<1	<1	5.8 ± 0.3

a. Concentration ± 2 sigma.

b. Composite of water from the #5 moisture cups of Lysimeters 1 through 4.

c. Moisture cup identity number.

d. Leachate collector identity number.

TABLE 8. RESULTS OF GAMMA-RAY AND STRONTIUM ANALYSES OF ORNL SOIL MOISTURE AND LEACHATE SAMPLES--JUNE 1986

Sample Identification	Concentration[a] (pCi/L)			
	Cobalt-60	Cesium-137	Antimony-125	Strontium-90
Composite[b]	<19	<19	<27	8.6 ± 10.2
Lys 1-3[c]	<27	<22	<27	0.5 ± 7.6
Lys 2-3	<81	<54	<27	15.4 ± 18.1
Lys 3-3	<54	<54	<27	32.4 ± 18.9
Lys 4-3	<27	<16	<27	<5
Lys 5-3	89.2 ± 32.4	<3	540 ± 81	17.6 ± 12.2
Lys 1[d]	<5	<5	<3	9.2 ± 3.5
Lys 2	<8	<6	<0.3	2.4 ± 2.7
Lys 3	<8	<6	<0.3	1.1 ± 2.7
Lys 4	<8	<5	<0.3	11.6 ± 4.6
Lys 5	<8	<5	<37	2.4 ± 2.9

a. Concentration ±2 sigma.

b. Composite of water from the No. 5 moisture cups of Lysimeters 1 through 4.

c. Moisture cup identify number.

d. Leachate collector identity number.

three times that of 5-3. In general, occurrence of ^{90}Sr in the leachate sample at both sites was down sharply from the April sampling, with measurable amounts being found only in ANL 5 and ORNL 1 and 4.

Occurrence of nuclides in the water samples from both the soil and inert sand lysimeters in such a short period of time (months rather than years) was unexpected. While ^{90}Sr is known to be soluble in soil solution and does move through the soil column almost unhindered by the soil matrix, it appears that leaching and movement of the nuclides is occurring at a more accelerated rate in the soil than was thought possible. The appearance of ^{90}Sr indicates that small quantities were readily leached from the waste forms. The higher initial concentration at ORNL could reflect the greater pore volume of water that has passed through these lysimeters. When comparing ^{90}Sr data from the April and June samplings, it appears that ^{90}Sr moved through the lysimeters as an initial slug which, in the case of ORNL, has been washed out or, as at ANL-E, is in the process of being flushed. Data from ANL 5 would support this hypothesis, since it appears that a plume of ^{90}Sr movement has been detected in this lysimeter, with the trailing edge showing up in the area of ANL 5-3, the bulk near ANL 5-1, and a leading edge moving into the leachate collector. Though ^{90}Sr has been detected, the total quantity leached is only a small fraction of that available in the waste forms (Table 9).

Finally, because it is apparent that the soil in the lysimeters has subsided (very evident at ANL-E), it was decided to determine if the movement had caused a shift in the position of the waste forms. This was accomplished by lowering a radiation-detecting probe down the access tube which leads into the leachate holding tank. Readings were taken every 15.2 cm in all lysimeters, and radiation intensity with depth was recorded. Readings of the soil lysimeters were then compared with readings from the sand-filled controls. At ORNL, the intensity of radiation readings for each lysimeter approximated the known depth of the waste forms (Table 10). However, at ANL-E, some settling has occurred; readings in the soil-filled lysimeters (1-4) were still elevated at the 182.9-cm depth, while the

TABLE 9. COMPARISON OF TOTAL STRONTIUM-90 PER LYSIMETER TO AMOUNT FOUND IN ORNL LEACHATE--APRIL 1986

Lysimeter Number	Waste Form Involved	Amount Sr-90 per Waste Form (pCi)	Total Sr-90 per Lysimeter[a] (pCi)	Sr-90 in Leachate (pCi/L)	Total Sr-90 Leached[b] (pCi)	Percent of Total SR-90 in Leachate
1	C1 + C1A cement + PF7 (high Sr-90)	2.64 E9	18.2 E9	62 ± 8	11.7 E3	.000064
2	C2B cement + PF24 (low Sr-90)	0.47 E9	3.3 E9	17 ± 5	5.1 E3	.000016
3	D1A Dow + PF7 (high Sr-90)	3.92 E9	27.4 E9	4.9 ± 3	0.9 E3	.000003
4	D2 Dow + PF24 (low Sr-90)	0.64 E9	4.5 E9	54 ± 8	10.2 E3	.000023
5	C2B cement + PF7 (low Sr-90)	0.47 E9	3.3 E9	46 ± 8	8.7 E3	.000026

a. Value calculated--seven waste forms x pCi content of waste form = total pCi/lysimeter.

b. Value calculated--pCi/L x number of L per total sample (≡ 189) = total pCi leached.

TABLE 10. RADIATION INTENSITY WITH DEPTH IN EPICOR-II FIELD LYSIMETERS

Depth from Soil Surface (cm)	Radiation Intensity (mR/h)									
	ANL-E Lysimeter Number					ORNL Lysimeter Number				
	1	2	3	4	5	1	2	3	4	5
15.2	--a	--a	--a	--a	--a	--a	--a	--a	--a	--a
30.5	--a	--a	--a	--a	--a	--a	--a	--a	--a	--a
45.7	--a	--a	--a	--a	--a	--a	0.02	0.04	0.01	0.04
61.0	--a	--a	--a	0.3	1.0	0.005	0.04	0.18	0.29	0.21
76.2	--a	--a	--a	1.0	3.0	0.16	0.24	0.65	1.6	1.4
91.4	0.4	--a	0.5	1.3	6.0	0.81	1.0	2.7	1.6	1.4
106.7	0.7	2.0	2.5	7.0	10.0	2.9	3.5	13.7	25.8	20.8
121.9[b]	3.5	18.0	5.0	35.0	12.0	6.1	11.2	29.0	52.4	40.3
137.2	6.0	28.0	20.0	43.0	12.0	11.2	16.1	39.5	70.2	48.3
152.4	7.5	39.0	18.0	67.0	12.0	11.2	17.7	40.3	70.9	36.3
167.6[b]	7.0	32.0	17.0	65.0	8.0	8.1	12.9	30.6	54.8	20.9
182.9	3.6	21.0	10.0	38.0	6.0	2.4	4.6	14.5	25.8	5.9
198.1	1.8	10.0	2.5	18.0	5.0	0.73	1.5	3.7	9.7	1.1
213.4	--a	--a	--a	--a	--a	0.16	0.31	0.89	1.4	0.16

a. Readings were not above background.

b. Location of waste form is indicated by sets of bars.

activity in the inert control had moderated by that depth, indicating a downward movement of about 7.5 cm in the soil-filled lysimeters. There is no evidence that this movement has impacted the experiment except for minor damage to some moisture/temperature probes.

CONCLUSIONS AND RECOMMENDATIONS

Lysimeter operation at ANL-E and ORNL has had a successful first year. Despite some operational problems, the detectors and DAS system have provided appropriate data for the production of a visual portrayal of the year-long environmental conditions experienced by each lysimeter. It was discovered that ^{90}Sr is moving from the waste forms into the soil column much sooner than expected. This movement was especially apparent at ORNL, where ^{90}Sr was found in all leachate samples. It is assumed that the characteristics of the ORNL environment and Barnwell-type soils caused more movement of ^{90}Sr at ORNL than ANL-E by allowing a greater quantity of precipitation to infiltrate and percolate through the soils. ANL-E however, had a persistent occurrence of ^{60}Co in water obtained from a moisture cup. Since cobalt is not "fixed" by soils (like cesium), its movement is expected to continue.

The NRC recommended that water samples be analyzed for ^{90}Sr at each sampling period. This suggestion appears to be worthwhile and was incorporated into the experimental work plan. After two samplings, ^{90}Sr has been identified in a number of lysimeters.

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A field study was designed to monitor the release (if any) of beta- and gamma-producing radionuclides from solidified EPICOR-II ion exchange resins. Both Portland Type I-II cement and Dow vinyl ester-styrene waste forms are being tested in lysimeter arrays located at Argonne National Laboratory in Illinois (ANL-E) and Oak Ridge National Laboratory (ORNL). The study is designed so that continuous data on nuclide release and movement, as well as environmental conditions, will be obtained over a 20-yr period. Results of the first year of data acquisition are presented and discussed. Unusual occurrences over that period are also described.

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